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High-temporal-resolution X-ray spectroscopy with free-electron and optical lasers

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Ultrafast X-ray spectroscopies require flexible X-ray properties together with high temporal and spectral resolution. Here, we demonstrate simultaneous sub-20 fs and sub-eV resolutions for pump/probe experiments, without the need for additional photon arrival-time monitors.

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High-energy photons in the X-ray spectral region can directly interact with inner electronic orbitals. The binding energies of these core electrons are representative of the parent atomic species because they strongly vary with the atomic number. However, despite the localized nature of these inner orbitals, changes in the environment surrounding the target atom can shift the respective binding energies by up to a few eV [1]. Resolving these chemical shifts via X-ray spectroscopies allows characterizing the local chemical environment of the individual atoms embedded in larger structures, a property known as site selectivity.

This property becomes particularly attractive when combined with femtosecond pump/probe spectroscopy to resolve photo-induced structural rearrangements that occur when nuclei react to changes in the electronic structure [2]. For such experiments, the temporal resolution is limited by the pump and probe pulse durations, and by their relative arrival-time jitter. So far, only X-ray free-electron lasers (XFELs) have been able to provide X-ray pulses with narrowband and broadly tunable spectra and femtosecond durations; however, their synchronization with a pump optical laser yields timing jitters of several tens of femtoseconds [3,4]. Further improvements can be achieved through independent photon arrival-time measurements [5], which, however, require specific X-ray fluences on the photon-arrival measurement device, constraining the operating parameter range of the FEL radiation on the actual experiment.

In this work, we demonstrate that using the dedicated European XFEL optical synchronization system [4,6], together with noninvasive arrival-time monitors for relativistic electrons [7,8] enables temporal resolutions below 20 fs FWHM without the use of additional photon-arrival measurements. The simultaneous use of an X-ray monochromator [9] enables a sub-eV spectral resolution without significantly compromising the achieved temporal resolution, for present FEL beam conditions.

The experiments were realized at the small quantum systems (SQS) instrument of the European XFEL. The SASE3 soft X-ray undulator was seeded by electron bunches of 14 GeV in energy and a charge of 250 pC. The undulator was tuned to provide X-ray pulses centered at a photon energy of 1320 eV with a 4 eV FWHM bandwidth and energies up to 300 μJ. Single-shot spectral measurements performed behind the SASE3 undulator revealed a group duration below 10 fs FWHM obtained through spectral correlation analysis [10]. The monochromator was used to reduce the FEL bandwidth to 0.3 eV FWHM [9].

Our proof-of-principle experiment is based on the laser-assisted X-ray photoemission technique [11]. The X-ray pulses ionized electrons from the Ne 1s shell (binding energy of 870.2 eV) and the photoelectrons were detected with a horizontal electron time-of-flight (eTOF) spectrometer, measuring parallel to the linear FEL polarization [12]. A retardation voltage of 420 V was used to detect the 450-eV kinetic energy photoelectrons.

To "dress" the photoemission process, the dedicated SASE3 OPCPA-based optical laser was used (similar to [13] and references therein). For this experiment, the system was set to provide linearly, vertically polarized pulses with a 15 fs FWHM duration and a central wavelength of 800 nm, producing sidebands around the main photoline, each separated by an energy equal to an integer number of optical photons (1.55 eV). With a laser peak intensity of 8.8×10^{13} W/cm^2 and an (almost) perpendicular polarization of the optical laser with respect to the horizontal eToF spectrometer, sidebands up to the fourth order were generated [14].

By varying the delay between the optical laser and the FEL, the temporal dependence of the laser-dressing effect was measured, which corresponds to an FEL/laser cross-correlation [11]. The

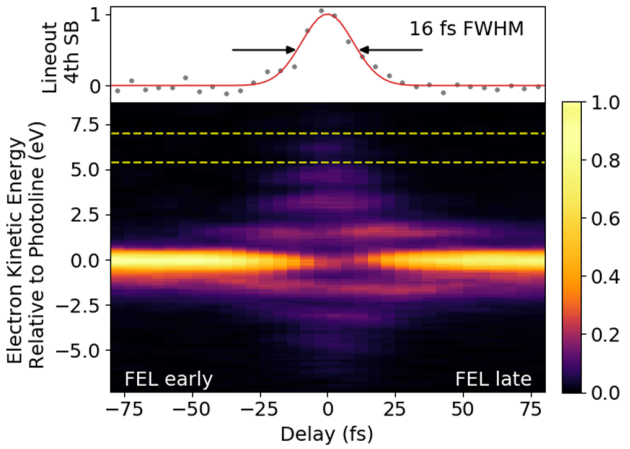

Fig. 1. Ne 1s photoelectron spectrum as a function of relative laser/FEL delay. The laser-dressing effect generates sidebands of the photoline at 449.8 eV separated by 1.55 eV. The projection of the fourth-order sideband marked by the yellow lines is fitted with a Gaussian of 16 fs ± 2 fs FWHM duration, which corresponds to a four-optical-photon/X-ray cross-correlation convoluted with the residual temporal jitter. The measurement consisted of 215,096 shots, using 290 pulses per second.

relative delay was obtained by varying the path length of the optical laser to the interaction region and correcting it by the arrival time of the electrons, measured for every shot by the bunch arrival-time monitor [8].

The resulting photoelectron spectrum plotted as a function of FEL-laser delay is shown in Fig. 1. As the FEL bandwidth is narrower than the photon energy of the optical laser, distinct sidebands of the photoline are observed when both the FEL and laser temporally overlap. The projection of the fourth-order sideband in time is fitted with a Gaussian function, retrieving a temporal width of 16 fs ± 2 fs FWHM (the error corresponding to the fitting error), defining the temporal resolution of the technique. Taking into account that this is a four-optical-photon process, this value is consistent with an X-ray pulse duration and residual temporal jitter of approximately 10 fs FWHM each. The photoline itself has a FWHM bandwidth of 0.92 eV, as obtained from delays below −60 fs where the laser dressing is absent. This value corresponds to a convolution between the X-ray bandwidth of 0.3 eV, the neon 1s core-hole lifetime bandwidth of 0.27 eV and an estimated spectrometer resolution of 0.83 eV at the specific settings used for this measurement.

In conclusion, we have demonstrated a time-resolved measurement of a laser-assisted X-ray photoemission exhibiting a temporal resolution of 16 fs ± 2 fs FWHM with X-ray pulses of 0.3 eV FWHM bandwidth. This was achieved without using additional photon arrival monitors and allows full flexibility on the properties of the X-ray beam. Although we benchmark these capabilities using soft X-ray photoelectron spectroscopy, they are general to other types of experiments, providing the possibility for high repetition-rate single-shot acquisitions for electron arrival-time correction.

To ensure stable operation over longer periods of time, further investigations are ongoing to actively minimize electron arrival-time jitter and long-term drifts. This also involves further investigation to quantify timing changes related to varying environmental conditions in the accelerator, the instrument, and on the optical laser delivery path. In addition, the use of high-resolution timing tools is envisioned [15], supporting temporal resolutions below the optical synchronization limit. This capability will become particularly important with the upcoming development of X-ray pulses with durations in the attosecond regime [16,17].

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Data availability. Data underlying the results presented in this Letter will be available in [18].

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